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Evidence of low levels of trace organic contaminants in terminal lakes

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ABSTRACT

Endorheic lakes (or terminal lakes, TLs) have no natural outlet other than evaporation and slow infiltration. Some TLs receive reclaimed wastewater which contains poorly removed trace organic contaminants (TrOCs). To determine if TLs accumulate TrOCs we conducted a preliminary assessment of the occurrence of ten TrOCs in three TLs receiving reclaimed wastewater and one TL which does not directly receive reclaimed wastewater. Five of ten TrOCs (carbamazepine, DEET, fluoxetine, primidone, and trimethoprim) were present in all four TLs' surface waters (~0.3–1109 ng/L), six (caffeine, carbamazepine, DEET, diphenhydramine, primidone, and trimethoprim) were present in sediment samples (0.1–77 ng/gDW) and in soil samples (0.1–137 ng/gDW). Concentrations of caffeine, carbamazepine, diphenhydramine, fluoxetine and meprobamate were significantly higher in TLs receiving wastewater from a secondary treatment plant compared to those TLs which received tertiary treated wastewater. Carbamazepine, fluoxetine, sulfamethoxazole, and trimethoprim were present at concentrations greater than is typical of other U.S. freshwater lakes, but other TrOC concentrations were present at lower concentrations than in other freshwater lakes. We conclude that some TrOCs may accumulate in TLs, but to a lesser extent than would be expected based on the accumulation of dissolved constituents alone, which indicates that there are other unidentified processes in TLs that contribute to TrOC losses. Other TLs across the globe may have similar levels of TrOCs due to anthropogenic influence and treated wastewater inputs.

1. Introduction

Endorheic lakes (commonly known as terminal lakes (TLs)), are hydrologically closed basins common in arid and semi-arid environments (Wurtsbaugh et al., 2017; Wang et al., 2018). Notable regions containing a high frequency of TLs include western and central Asia, north Africa, central Australia, and the Great Basin of North America (Yapiyev et al., 2017). TLs receive water from surface water flow and precipitation and have only two outlets: evaporation and groundwater infiltration. TLs are important habitats to various endemic species (Larson, 2014; Schrage, 2016; Sorensen et al., 2020; Wurtsbaugh et al., 2017).

One of the major threats in the last several decades that TLs face globally is increasing salinity and total dissolved solids due to water diversion and climate change causing reduced precipitation and increased evaporation (Timms, 2005; Wang et al., 2018). Increased salinity jeopardizes the survivability of species such as the threatened Lahontan cutthroat trout, endemic to TLs and streams of northern Nevada, northern California, and Oregon (Dickerson and Vinyard, 1999; Herbst et al., 2013; Sedinger et al., 2012; Wagner et al., 2001).

One method to replace inflow losses is to augment flows with treated wastewater but treated wastewater contains trace organic contaminants (TrOCs) (Sharma et al., 2020); pharmaceuticals and personal care products, endocrine-disruptors, and other organic synthetic compounds (Dodgen et al., 2015; Goldstein et al., 2014; Pan et al., 2016; Wu et al., 2014; Zessner et al., 2020). TrOCs may also enter TLs through overland flow (e.g., atrazine) or shoreline anthropogenic activity. Exposure to TrOCs present in treated wastewater is known to proliferate antibiotic resistance, disrupt the endocrine system (e.g. reduce fertility, feminization of males, and intersex phenomena) (Richardson et al., 2005) and cause mode of action driven effects in fish (e.g. consuming abnormality, predator abstention and courtship display) (Geyer et al., 2000; Kemper, 2008; Tanoue et al., 2015).

TrOCs have varying environmental fate and transport processes such as biodegradation, sorption, photodegradation, volatilization, oxidation, and hydrolysis, ultimately leading to their reduced aqueous concentrations (Wilkinson et al., 2017; Yi et al., 2020). Multiple published studies have examined the occurrence of TrOCs in streams, freshwater lakes, and oceans/seas (Kolpin et al., 2002; Meyer et al., 2019; Schultz et al., 2010; Yang et al., 2020; Zhang et al., 2007). For example, caffeine,

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carbamazepine and primidone were present in a river influenced by wastewater at concentrations between 2 and 687 ng/L (Guo and Krasner, 2009), and, caffeine, metformin, sulfamethoxazole, and triclosan were frequently detected in surface water and sediments of Lake Michigan (Blair et al., 2013; Ferguson et al., 2013). Several other PPCPs were also present in marine sediments in the ng/gDW range (Elliott et al., 2017; Long et al., 2013).

While there is a significant amount of data published on TrOCs in surface water, this data disproportionately focuses on lotic compared to lentic systems (Meyer et al., 2019), and we are unaware of any published literature on TLs. Given that 25% of the non-polar continental area of the earth is endorheic (Galat et al., 1981; Hostetler et al., 1995), we believe it is important to understand the fate of TrOCs in these systems. Our objectives were to a) determine the concentrations of ten TrOCs in TLs influenced by reclaimed wastewater and compare their concentrations to a TL which was not influenced by wastewater, b) compare TrOC concentrations in TLs to other U.S. lakes published in the literature and c) determine TrOC concentrations in shoreline soils vs nearshore sediments at each TL to investigate the impact of shoreline recreational activity and overland flow. Our hypotheses were that 1) TrOCs are present at higher concentrations in the lake than the lake influents due to accumulation of dissolved constituents in the lakes, 2) lake sediment concentrations are less than shoreline soils, and 3) TrOCs in TLs are present at greater concentrations than other U.S. lakes. We intentionally limited the scope to single sampling events of each lake and did not specifically investigate mechanisms of loss because we intend this study to act as a first screening of TrOCs in TLs and provide evidence for further investigation.

2. Materials and methods

2.1. Chemicals

Ten compounds were selected based on their frequent detections in wastewater treatment effluent and based on established in-house extraction and analysis methods (Sharma et al., 2020). Caffeine (CAF), carbamazepine (CBZ), *N,N*-diethyl-*meta*-toluamide (DEET), diphenhydramine (DPH), fluoxetine (FLX), meprobamate (MPB), primidone (PMD), trimethoprim (TMP), atrazine-d_5, caffeine-d_3, carbamazepine-d_{10}, diphenhydramine-d_3, and meprobamate-d_3 were purchased from Sigma-Aldrich (St. Louis, MO). Atrazine (ATZ) and fluoxetine-d_5 were purchased from Cayman Chemicals (Ann Arbor, MI). Sulfamethoxazole (SMX) was obtained from MP Biomedicals (Solon, OH). DEET-d_{10}, primidone-d_5, sulfamethoxazole-d_4, and trimethoprim-d_3 were purchased from Santa Cruz Biotechnology (Dallas, TX). Methyl tertiary-butyl ether (MTBE), methanol, HPLC grade water, diatomaceous earth, 0.1% formic acid (v/v) in water and 0.1% formic acid (v/v) in acetonitrile were acquired from Thermo Fisher Scientific (Waltham, MA). All analytical solvents and standards were of ≥98% pure except ammonium acetate, ≥97%, purchased from VWR International, LLC (Solon, OH).

2.2. Sample collection

Four TLs' surface waters, their nearshore sediments and shoreline soils in the U.S. state of Nevada were sampled once in the Fall of 2018 and Spring of 2019. Three to four locations at each lake were chosen based on accessibility. TL1, TL2, and TL3 are influenced by reclaimed wastewater. One TL does not receive reclaimed wastewater (CTL). For each lake, sampling location A is at the mixing zone with the inflow. To account for spatial heterogeneity, sediment and soil sampling locations B, C, and D were along the length of the shoreline of the lake and data were combined. ~20% of the collected samples were collected in triplicate.

Surface water samples were collected at least 2 m from the shore with a HDPE container attached to a pole. Samples were immediately transferred into 1 L amber glass bottles which had been washed, baked at 500 °C for 3 h, and preserved with 50 mg of ascorbic acid and 1 g of sodium azide.

Sediments were sampled with a stainless-steel hand auger from the lakebed, approximately 2 m from the shore, generally at a depth of ~10 cm. For sampling sites with more gravel which filled the hang auger, samples were collected at a depth >10 cm but less than 20 cm, where fine sediments were observed.

Surface soil samples (0–5 cm) were collected with a trowel. Soils were sampled from areas with minimal or no interaction with the lake water to determine the direct impact of human activity on the soil. The soil samples from all TLs were sampled from locations where the lake water does not interact with the soil but is near the surface water/sediment sampling locations. Only a single soil sample at location A of TL1 was sampled as other soil areas were inaccessible.

All sampling containers and tools were rinsed with DI water three times before and between samplings to minimize contamination. The trowel and hand auger were also rinsed and wiped with a paper towel between samples. Samples were transferred on ice to the University of Nevada, Reno laboratories. Soil and sediment samples were stored at −20 °C until analysis. Water samples were stored at 4 °C and typically analyzed within 2 weeks.

2.3. TrOC concentrations in U.S. freshwater lakes from literature

To compare TrOCs concentrations in TLs with other U.S. lakes, we searched published peer reviewed literature using the keywords pharmaceuticals and TrOCs in lakes, streams, rivers, and lake sediments in Harzing's Publish or Persing software (Harzing, 2007) to extract articles from Google Scholar using the method by AS Pullin et al. (2018). From the results we selected 9 publications that investigated the same TrOCs in the U.S. lakes as in this study. We also aggregated data from U.S. rivers and streams (n = 12) but limit our comparisons because these are flowing systems and are not expected to accumulate dissolved constituents. We also limited our comparison of TrOC concentrations in U.S. TLs to U.S. freshwater lakes because wastewater concentrations of TrOCs vary significantly across countries. A PRISMA diagram with the search results is included in Figure S1. TrOC concentrations extracted from the literature are compiled in Table S1 (Blair et al., 2013; Brent et al., 2001; Elliott et al., 2017; Ferguson et al., 2013; Ferrey et al., 2015; Boyd and Furlong, 2002; Lee et al., 2015; Tierney et al., 1999; Wang et al., 2020) and the data from this study is in Table S2.

2.4. Description of sampled lakes

TL1: Relatively shallow with a ~2 m maximum depth, 4 km^2 of surface area, and dominated by peri-urban surface runoff. Being a shallow lake, this lake does not go through turnover. The lake also receives reclaimed wastewater from two wastewater treatment plants via outfalls, one has biological treatment with disinfection and the other has tertiary treatment processes (sand filtration). The biological and tertiary treatment plants have flow rates of <0.04 m^3/s and discharges treated wastewater from 0.02 to 0.11 m^3/s respectively. Samples were collected on March 29th, 2019. One surface water, sediment and soil sample were collected from the mixing zone where the wastewater treatment plant's effluent canal mixes with the lake (location A). Only one soil sample was collected because other soil sample locations were strongly influenced seasonally by lake water and the lake is not regularly used for shore recreation. Overall, three surface water and sediment samples were collected at TL1.

TL2: Maximum depth of ~4 m with surface area of ~1 km^2 and receives water from a river which receives tertiary treated wastewater from a wastewater treatment plant. The river has an average annual flow rate of 11 m^3/s. The wastewater treatment plant flow averages 0.22 m^3/s. The plant consists of primary treatment, aerated biological treatment, filtration, and disinfection. Four surface water, sediment, and soil

samples were collected on March 23rd, 2019. This TL has a moderate level of shore recreation (e.g., hunting).

TL3: ~91 m deep with surface area of 490 km^2 and receives water from a river which is influenced by reclaimed wastewater (tertiary treatment with sand filters). This treatment plant treats an average flow of 1.36 m^3/s and discharges approximately 1.2 m^3/s to the river. The river has an annual average flow rate of 23 m^3/s. Four surface water, sediment and soil samples were collected on November 29th, 2018. This TL has a relatively high level of shore recreation (e.g., bathing, fishing).

CTL: ~152 m deep with 130 km^2 of surface area. This lake receives water from a river which is not impacted by reclaimed wastewater. Three surface water, sediment and soil samples were collected on May 8th, 2019. Annual average flow of the river is 4.6 m^3/s, some of which is diverted for irrigation prior to the lake.

WW effluents were not collected during sampling because the associated wastewater treatment plants are not easily accessed by the public and effluent releases are offset from lake influent by unknown or poorly understood time delays. The names and locations of the lakes are intentionally concealed because one lake lies on nonpublic land. The sampled lakes are relatively remote and poorly studied. Thus, little or no additional information related to evaporation and infiltration rates exists in published literature.

2.5. Sample extraction and analysis

The aqueous, soil and sediment samples were extracted and analyzed using methods described previously (Sharma et al., 2020). Briefly, aqueous samples were filtered with a 0.7 μm microfiber glass filters and 10 isotopically labeled standards were spiked to make the sample concentration equal to 100 ng/L of each labeled standard. Oasis Hydrophilic Lipophilic Balance (HLB) 6 cc/200 mg cartridges were preconditioned sequentially with 5 mL of MTBE, methanol, and ultrapure water, samples were then loaded to cartridges by an automated solid phase extraction system, the cartridges were dried under N_2 gas, and eluted with 5 mL of methanol followed by 5 mL of 10/90 (v/v) methanol/MTBE solution. The eluate was evaporated to near dryness under nitrogen gas flow, reconstituted to 1 mL with methanol, and stored at −20 °C until analyzed. Analysis of all compounds was conducted by liquid chromatography-tandem mass spectrometry (LC/MS-MS) in positive electrospray ionization (ESI+) mode.

Soil and sediment samples were freeze-dried and extracted by pressurized liquid extraction (ASE 200). Briefly, the freeze-dried sample was packed into extraction cells with diatomaceous earth used as filler. The extraction solvent was 2:1 v/v water-methanol solution at 100 °C and 1500 psi, and extraction was conducted three times. The extract was diluted to 1 L with ultrapure water. Solid phase extraction with HLB cartridges and elution with methanol was conducted as a clean-up method. Eluates were dried, reconstituted to 1 mL with (2:1, v/v) water-methanol solution, spiked with isotopes and stored at −20 °C until measurement by LC/MS-MS.

Quantification was achieved following the analytical method described by Sharma et al. (2020). Briefly, a Thermo Scientific UltiMate 3000 UPHLC with an Agilent RRHD ZORBAX Eclipse Plus reverse phase C18 column (2.1 × 50 mm, and 1.8 μm) was used for LC. The mobile phase was ultrapure water with 0.1% formic acid (solvent A) and acetonitrile with 0.1% formic acid (solvent B) with flowrate of 400 μL/min at 30 °C in ESI+ mode. The Thermo Scientific triple quad MS was tuned daily with caffeine and one ion transition was used for quantitation of each compound. Another transition was used for qualification. MS/MS parameters, analyte reporting limits and LC-MS/MS ion transitions are provided in Table S3, S4, and S5, respectively. Analytical check standards were within ±20% of their known concentration. Method reporting limits ranged from <1 ng/L in the aqueous samples to 10 ng/g in solids.

2.6. Statistical methods and data reporting

ANOVA with post-hoc Tukey's HSD was conducted in R v3.63 using the aov and TukeyHSD functions (R Core Team, 2019). The threshold for statistical significance was set at $p = 0.05$.

To reduce the amount of data shown in Figs. 1–3, we have shown only data above the reporting limit (see SI), but in the text have discussed all measurements above the quantification limit.

3. Results and discussion

3.1. TrOC occurrence in TLs

Ten TrOCs were quantified in the surface water of four TLs and the results are presented in Fig. 1. Surface water enters from location A into each TL, and locations B, C & D are along the lake shore between 1.1 km and 6.4 km from A. Nine TrOCs were detectable in TL1, more than any other lake. TL1 receives wastewater from two WW facilities, one is highly treated reclaimed wastewater, and the other treatment facility discharges to evaporation ponds which are directly adjacent to the lake. During the sampling event, the lake level was extremely high, causing localized flooding, and the evaporation ponds likely commingled with the lake water to some extent. CBZ, DEET, PMD and TMP were in all four lakes from 1 ± 0.6 (CBZ) to 1109 ± 1887 ng/L (PMD).

Sediment TrOC concentrations are in Fig. 2. Six TrOCs (CAF, CBZ, DEET, DPH, PMD and TMP) were in all four lakes from 0.15 ± 0.10 (DEET in TL1) to 12.40 ± 5.2 ng/gDW (TMP in TL1). Primidone at location B/C/D in TL1 was at 77 ng/gDW, the highest TrOC concentration observed in all sediment samples. ATZ and MPB were not detected in any sediments collected. FLX and SMX ranged from below the detection limit (BDL) to 2 ng/gDW. Overall, TL1 surface water and sediments had a greater number of TrOCs compared to other lakes (Figs. 1 and 2).

Soil samples were collected to understand the effects of shoreline recreational activities (Fig. 3). Six TrOCs (CAF, CBZ, DEET, DPH, PMD and TMP) were at quantifiable levels in all TLs from 0.1 ± 0.15 to 137 ± 195 ng/gDW (CAF). CAF was at the greatest concentration of 361 ng/gDW in TL3 soil at location C. This specific soil sampling location is a relatively high traffic recreational beach. Similarly, at high public traffic location A, CBZ in CTL was at 133 ng/gDW. Other compounds were observed at low concentration such as FLX and MPB were at 0.1 ng/g DW and 1 ng/gDW at TL3, respectively, whereas SMX was at ~1 ng/

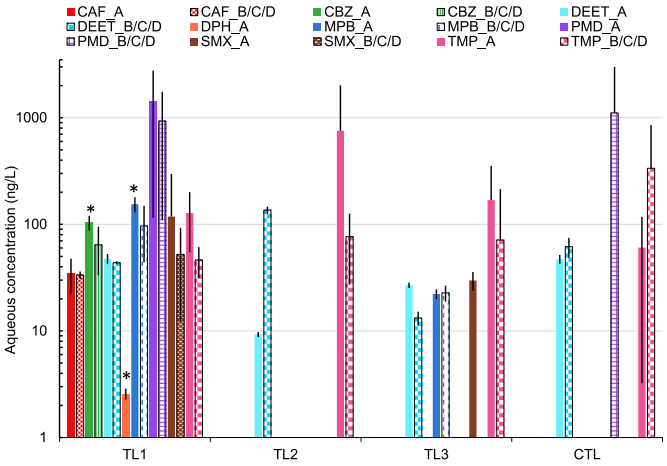

Fig. 1. TrOCs in the mixing zone (A, represented by solid bars) and the average of three nearshore sampling sites (B/C/D, pattern fills). Error bars show the standard deviation of three or more samples collected at location A and B/C/D of TL1, TL2, TL3 and CTL. Asterisks indicate the concentration is significantly different in the lake compared to the mixing zone. No bar indicates the analyte was below the reporting limit.

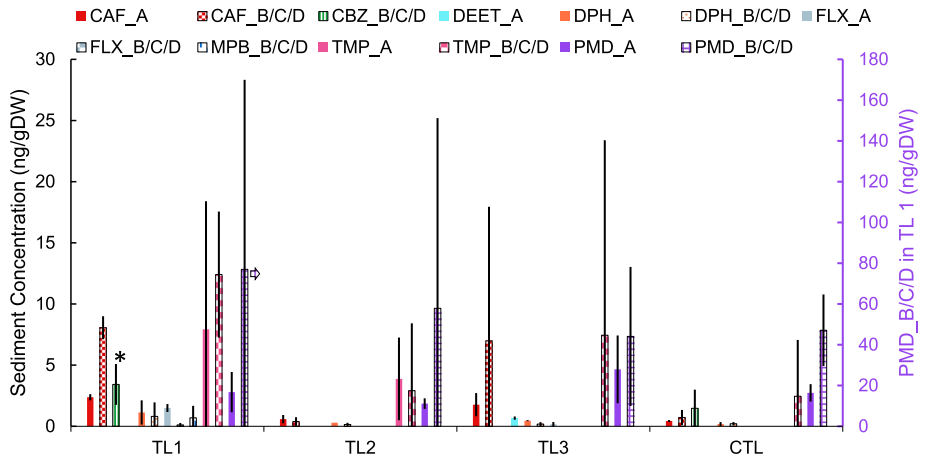

Fig. 2. TrOCs in sediments at the mixing zone (A) and the average of three points near the lakeshore (B/C/D). Error bars represents the standard deviation among three or more samples. Asterisks indicate the concentration is significantly different in the lake compared to the mixing zone. No bar indicates the analyte was below the reporting limit.

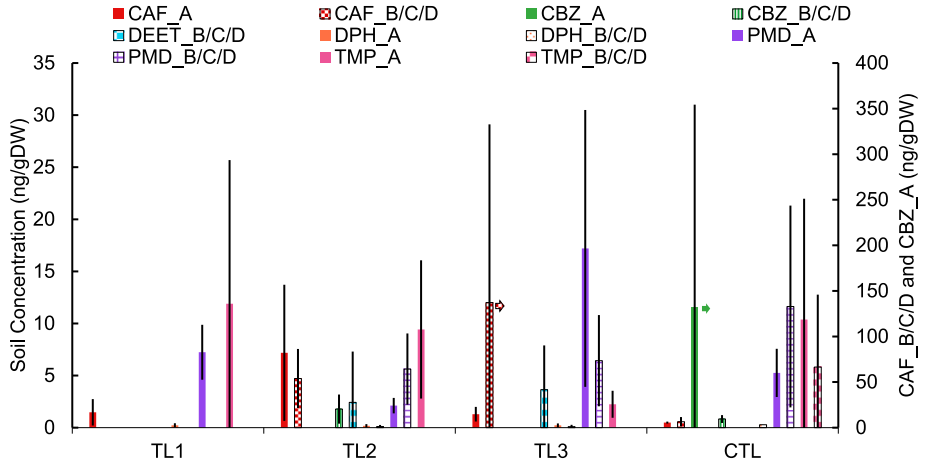

Fig. 3. TrOCs in soils at locations A & B/C/D. Locations B, C & D are the average of three TLs and one control lake. Error bars show the standard deviation of three or more samples. No bar indicates the analyte was below the reporting limit.

gDW at TL2 and TL3 soil samples. ATZ was below the detection limit in soil samples at all four lakes.

3.2. Variability among TLs

ANOVA was used to determine differences between samples (Table S6 and S7). CAF, CBZ, DPH, and MPB were in TL1 at greater concentrations than TL2, TL3 and CTL. TL1 receives wastewater with the lowest level of treatment and therefore the greater concentration of TrOCs was somewhat expected. CAF was not detected in TL1 but was in all other three lakes. There was no significant difference in concentrations of CBZ, DPH, MPB, PMD, SMX and TMP between TL2, TL3 and CTL. DEET and FLX were present at lower concentrations in both TL2 and TL3 compared to the CTL lake, even though TL2 & TL3 receive tertiary treated wastewater and CTL does not receive reclaimed wastewater. However, the concentration of DEET was greater in TL2 than TL3, potentially because TL2 is shallower than TL3 (i.e., less dilution), and, because of greater recreational activity by those who use insect repellent at TL2 compared to TL3 (i.e., TL2 attracts a large number of sportsmen/sportswomen). Overall, concentrations of TrOCs were similar in lakes with direct reclaimed wastewater input to those in a lake without wastewater input. For the lake with both reclaimed wastewater input and only biologically treated wastewater input, concentrations were significantly elevated.

Similarly, for sediments, DPH and FLX concentrations were greater in TL1 than TL2, TL3 and CTL but DEET was lower in TL1 than TL2, TL3 and CTL. The concentration of CAF, CBZ, PMD, SMX & TMP were not significantly different between the lakes but CAF, PMD and TMP were at modestly greater concentrations in TL1 sediments compared to the other three lakes. MPB was below the method reporting limits for all samples.

Comparing lake sediment to shoreline soils, we find that TL1's soil TrOC concentrations were either similar to or less than respective lake sediments. However, at every sampling location (A/B/C/D) of TL2, soil CAF concentrations were greater than sediments concentrations. Also at TL2, the concentrations of CBZ at location B, DEET at location D, and TMP at location A were greater than their respective sediment concentrations. Further, in TL3, CAF, DEET and PMD and in CTL, CBZ, PMD and TMP, were at higher concentration in soil at some sampling locations compared to respective sediment samples. There was no significant difference in TrOCs concentrations in soil samples of TLs impacted by reclaimed wastewater compared to the CTL lake. The lakes in this research all have some level of public shoreline activity based on local knowledge of boat ramps, camping locations, sports/hunting clubs, and non-quantitative observations made while sampling. This is well demonstrated by the intermittently greater concentrations in respective soils compared to sediments, and indicates that at least some of the TrOCs present in the lakes originated in shoreline activity rather than wastewater flows.

3.3. Losses in the TLs

We expected that concentrations of TrOCs would be greater in the lake compared to the mixing zone due to the accumulation of dissolved constituents. For CAF and DEET in TL2, PMD in TL3, and PMD and TMP in CTL, this was true; concentrations were greater at location B/C/D than location A in surface water samples. However, for TL1, the mixing zone (Location A) had significantly greater concentrations of CBZ, DPH, FLX & MPB than location B/C/D (Fig. 1). In TL2, TL3 and CTL the TrOC concentrations at locations B/C/D of surface water samples were not significantly different from their respective mixing zones. Thus, it is possible that sediment sorption and biodegradation and/or photolysis mediate TrOCs in the lakes, resulting in concentrations that are not significantly amplified by evaporative lake water losses (Cantwell et al., 2018; Yi et al., 2020).

This is further supported by sediment concentrations where TrOCs tended to be at higher concentrations in lake sediments than in the mixing zone sediments. This is most apparent for CAF and PMD, which were significantly greater in lake sediment than mixing zone sediment. Other TrOCs had an overall similar trend but differences were not statistically significant.

3.4. Comparisons of TLs with U.S. surface waters

TrOC concentrations in the U.S. lakes/rivers/streams and sediments published in the scientific literature were compiled and are in Figs. 4 and 5. The aggregated data is presented in Table S1. Comparisons here are made between TLs in this study and lakes with natural discharges to surface water (literature) because no data exists for TLs but also because we believed TLs may accumulate TrOCs. Comparisons between TLs and U.S. surface waters were variable, with some TrOCs at greater concentrations in TLs and some at lower concentrations. Average concentrations of ATZ, CAF, and DPH were at least four times greater in the U.S. lakes reported in the literature compared to the four TLs. DEET concentrations in the literature were greater by an average of 18 ng/L compared to TLs measured in this study. However, CBZ, FLX, and SMX were in TLs at concentrations approximately double that of the literature and TMP approximately 50 times higher than literature. PMD was also at high concentration in TLs but we are not aware of any similar literature with which to compare.

Concentrations of TrOCs that were above the reporting limits in sediment samples in this study and in literature, were 5, 55 and 71 times greater in freshwater lakes than in the TLs (CAF, DPH and FLX, respectively) (Blair et al., 2013; Lee et al., 2015). CBZ, DEET, SMX and TMP were either below the detection limits or no comparable data was available (PMD) in published literature. In this study, DEET, ATZ, MPB and SMX were below the detection/reporting limits and therefore were also not compared with literature.

Overall, CBZ, FLX, SMX and TMP were at higher concentrations and ATZ, CAF, DEET and DPH at lower concentrations in the aqueous phase of TLs compared to freshwater. ATZ, CAF, DPH and FLX were at lower concentrations in TLs sediments than freshwater lake sediments. TMP was at higher concentration in TLs sediments. These differences highlight the variable mechanisms of degradation and transport of TrOCs from surface water to sediment. We note that the samples investigated in this study represent only two seasons in a single 12 month period but believe the data is sufficient to demonstrate that TLs do not significantly accumulate TrOCs.

4. Conclusions

Some TrOCs (CBZ, FLX, SMX and TMP) were present at higher concentration in multiple TL surface waters compared to their concentrations in other U.S. lakes, but generally concentrations were less than would be expected based on the accumulation of dissolved constituents alone, indicating that degradation in the lake or lake sediments play a key role in TrOC losses. There was no significant differences in TrOC concentrations in TL soils near lakes impacted by reclaimed wastewater compared to those which were not wastewater impacted. In some sampling locations, TrOC concentrations were greater in soils compared to their respective sediment samples, potentially due to shoreline human activity. PMD was ubiquitous in aqueous and sediment samples, reflective of its relatively high anthropogenic use and long environmental lifespan. Future research should focus on the temporal nature of wastewater influence in the lakes, where during late summer wastewater flows would be expected to contribute significantly more than snowmelt/runoff. Another opportunity for future research is to investigate the mechanisms of TrOC loss or decomposition, as this may have treatment and management implications for more contaminated systems.

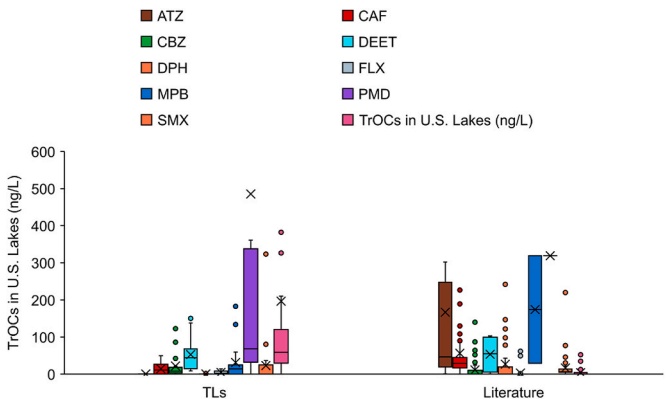

Fig. 4. Comparison between TrOCs in surface water of TLs (this study, n = 4) and literature (ranging from n = 1 to 6). Median shown by line, box shows interquartile range, cross is mean and individual points are outliers. Outliers (TLs PMD- 1128 ng/L, 1739 ng/L, 2901 ng/L and 3932 ng/L and Literature CAF- 800 ng/L) are not shown.

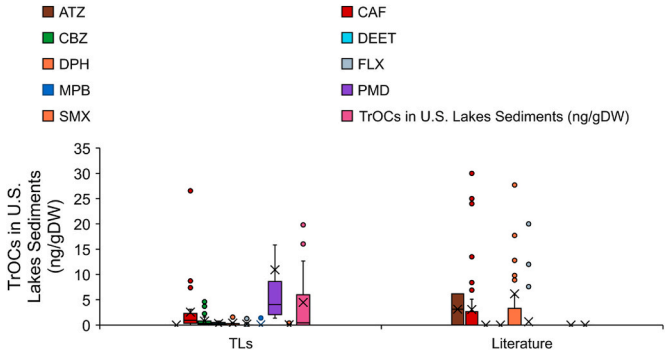

Fig. 5. TrOCs in sediments of TLs (this study, n = 4) and other U.S. lakes (literature, ranging from n = 1 to 3). Outliers (TLs PMD- 37 ng/g DW, 143 ng/gDW and Literature DPH- 36 ng/gDW, 37 ng/gDW, 44 ng/gDW, 48 ng/gDW, 82 ng/gDW) are not shown.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.131408.

Credit author statement

Priyamvada Sharma: Methodology, Software, Validation, Formal analysis, Investigation, Writing – original draft, Data curation, Visualization, David Hanigan: Conceptualization, writing-review and editing, Resources, Visualization, Supervision, Project administration, Funding acquisition

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